

[54] PRESSURE SENSITIVE RECORDING SHEET

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[73] Assignee: General Company Limited, Tokyo, Japan

[21] Appl. No.: 963,747

[22] Filed: Nov. 27, 1978

Related U.S. Application Data

[63] Continuation of Ser. No. 736,536, Oct. 28, 1976, abandoned, which is a continuation of Ser. No. 589,877, Jun. 24, 1975, abandoned.

[30] Foreign Application Priority Data

Oct. 31, 1974 [JP] Japan 49/124589
Jan. 31, 1975 [JP] Japan 50/12421
Jan. 31, 1975 [JP] Japan 50/12422

[51] Int. Cl.³ G11B 5/62

[52] U.S. Cl. 428/40; 346/77 E;
346/77 R; 346/135.1; 428/354; 428/422;
428/908; 428/913; 346/76 R; 77 R; 77
E; 135.1; 136; 137; 138

[58] Field of Search 428/908, 913, 40, 510;
428/571, 516, 517, 519, 520, 522, 354, 422;
40/2.2; 264/176 R, 127; 156/244

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Primary Examiner—Stanley S. Silverman

Attorney, Agent, or Firm—Fitzpatrick, Cella, Harper & Scinto

[57] ABSTRACT

A pressure sensitive member comprises a base sheet and an opaque pressure clarifiable layer overlying the substrate and the color of the substrate being different from that of the opaque clarifiable layer.

9 Claims, 19 Drawing Figures

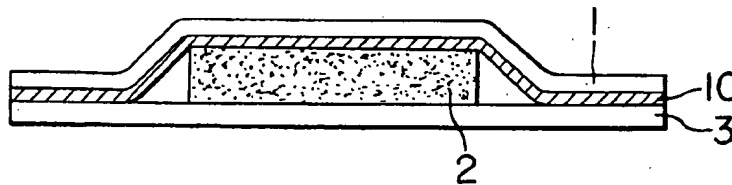


FIG. 1A

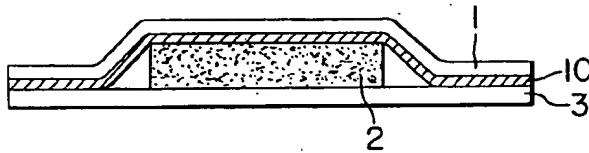


FIG. 1B

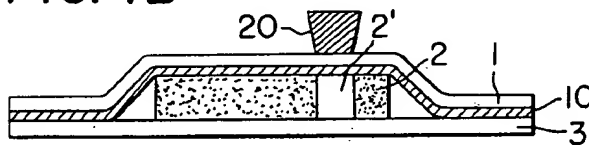


FIG. 2

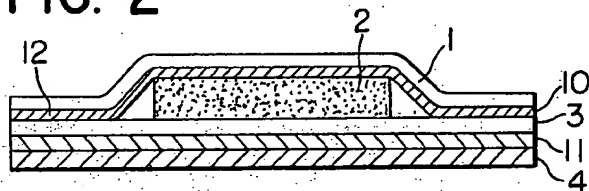


FIG. 3

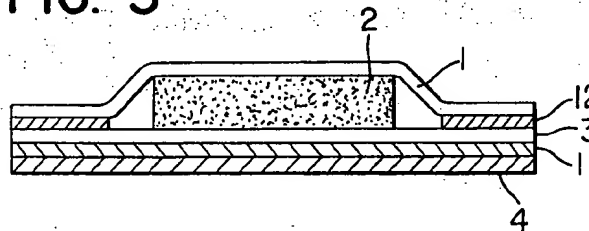


FIG. 4

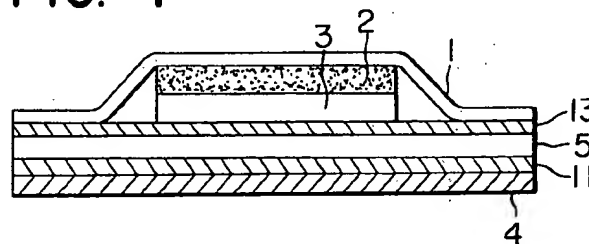


FIG. 5



FIG. 6

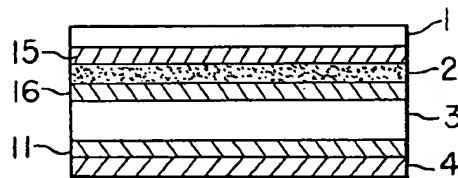


FIG. 7

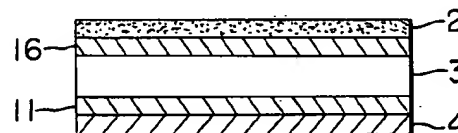


FIG. 8

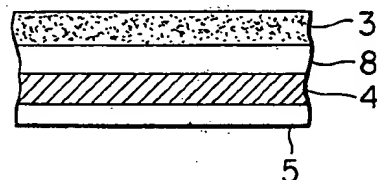


FIG. 9

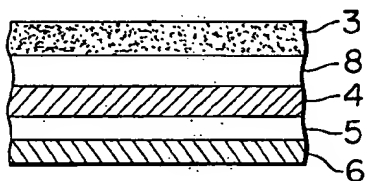


FIG. 10

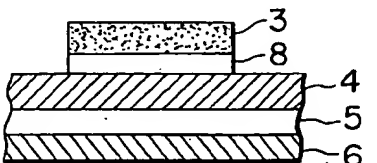


FIG. 11

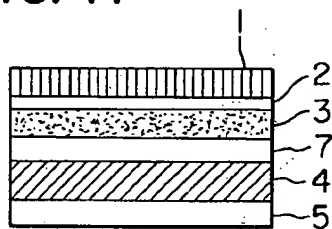


FIG. 12

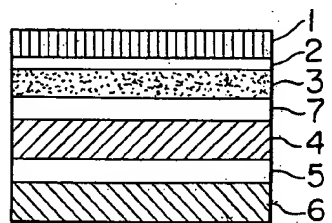


FIG. 13

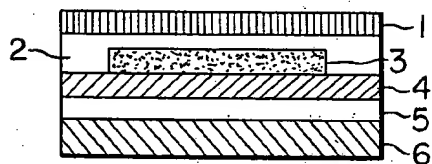


FIG. 14

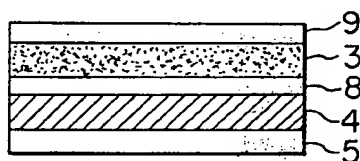


FIG. 15

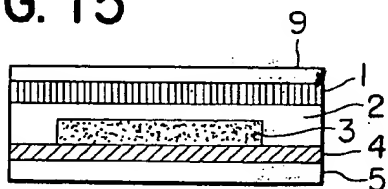


FIG. 16

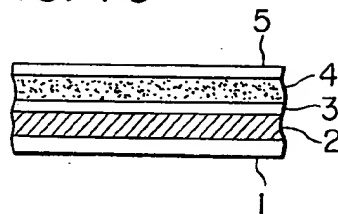


FIG. 17

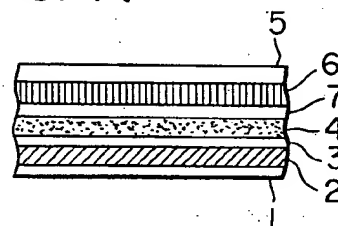
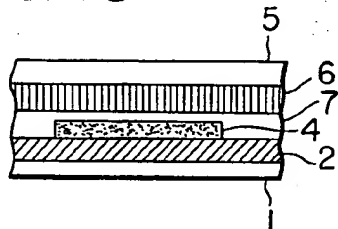


FIG. 18



PRESSURE SENSITIVE RECORDING SHEET

This is a continuation of application Ser. No. 736,536, filed Oct. 28, 1976, now abandoned, which is a continuation of application Ser. No. 589,877, filed June 24, 1975, now abandoned.

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to a pressure sensitive recording sheet.

2. Description of the Prior Art

There have been heretofore known inscribable labels. The simplest ones are those made of paper, but are of very poor water resistance and can not be used outdoors or in water. Another type of inscribable label is composed of a colored plastic base sheet, a crystalline wax coated thereon and a transparent film overlying the crystalline wax coat and the inscription can be effected from the transparent film side. However, this label has poor heat and light resistances and is not suitable for a long time use outdoors. A further conventional inscribable label is a so-called "embossing tape" composed of a colored backing sheet and a transparent film overlying the backing sheet which becomes whitened when subjected to deformation by a punch-die. The embossing tape has an excellent circumstance resistance. However, the inscription can not be made by a usual stylus or ball-point pen, but only by a special tool such as a punch-die.

SUMMARY OF THE INVENTION

According to one aspect of the present invention, there is provided a pressure sensitive recording sheet which comprises an opaque pressure clarifiable layer and a support sheet under the opaque pressure clarifiable layer, the support sheet having a color contrasting with the color of the opaque pressure clarifiable layer.

According to another aspect of the present invention, there is provided a pressure sensitive recording sheet which comprises the layer structure as mentioned above and additionally a transparent protective layer is provided on the opaque pressure clarifiable layer.

According to a further aspect of the present invention, there is provided a pressure sensitive recording sheet which comprises an opaque pressure clarifiable layer, a transparent support sheet under the opaque pressure clarifiable layer, and a colored adhesive layer under the transparent support sheet, and if desired, a transparent protective layer is provided on the opaque pressure clarifiable layer.

An object of the present invention is to provide a pressure sensitive recording sheet having excellent heat resistance, light resistance and water resistance, free from formation of crack and capable of being inscribed with a usual writing instrument.

Another object of the present invention is to provide a pressure sensitive recording sheet which can be easily produced.

BRIEF DESCRIPTION OF THE DRAWING

FIG. 1A and FIGS. 2-FIG. 18 diagrammatically show enlarged cross sectional views of embodiments of the pressure sensitive recording sheet according to the present invention; and

FIG. 1B diagrammatically shows that an inscription to a pressure sensitive recording sheet of the present invention.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Support sheets used for the present invention may be flexible or non-flexible materials and transparent or opaque depending upon the use. Further, the shape is not critical, but usually such forms as thin sheets, foils and films are preferable.

Representative support sheets include papers such as kraft paper and the like, plastic films such as acetylcellulose, polypropylene, polyethylene, polyester, and soft or hard polyvinyl chloride films and the like, and metal foils such as aluminum foil, copper foil and the like.

Opaque pressure clarifiable layers used in the present invention are layers which are opaque at an ordinary state and transparentize when a pressure is applied to them. These materials transparentize when a pressure such as that of a writing instrument and that of typewriter printing heads is applied to them.

Representative materials for the opaque pressure clarifiable layers are polyfluoroethylenes such as polytetrafluoroethylene and the like, and polyethylene such as low pressure process polyethylene and the like. There is preferably used unbaked polytetrafluoroethylene. This is a polytetrafluoroethylene as shaped by a paste extruding method and not followed by a heating treatment.

The transparent protective layer used in the present invention may be a transparent sheet, for example, film, which can transfer pressure patterns applied thereto to the opaque pressure clarifiable layer under the transparent protective layer and can protect the opaque pressure clarifiable layer from, for example, dirt, scratching and other damages. The transparent protective layer may serve for controlling the pressure sensitivity of the pressure sensitive recording sheet.

Representative materials for the transparent protective layer include thin transparent acetylcellulose, polypropylene, polyethylene, polyester, and soft or hard polyvinyl chloride films.

The pressure sensitive recording sheet comprising an opaque pressure clarifiable layer, a support sheet and, if desired, a transparent protective layer being provided on the opaque pressure clarifiable layer according to the present invention may have an adhesive layer under the support sheet, or both of an adhesive layer under the support sheet and a release treated layer on the top of the pressure sensitive recording sheet, that is, on the opaque pressure clarifiable layer when no transparent protective layer is mounted thereon, or on the transparent protective layer. Further if desired, a release layer may be provided under the adhesive layer. When the release layer is provided under the adhesive layer, the release treated layer on the top of the pressure sensitive recording sheet is usually unnecessary.

As the adhesive layers, there may be used usual adhesive materials such as, for example, pressure sensitive adhesives and water soluble adhesives. Representative pressure sensitive adhesive layers may be produced by dissolving rubbers such as neoprene, vinyl resins such as polyvinyl chloride, high polymers of cellulose series such as ethyl cellulose, together with an adhesivity imparting agent such as D.O.P., dammar and the like, in a solvent such as, for example, solvent naphtha, applying to the support sheet, and drying.

The release treated layer may be produced by conventional method for producing a release coat, for example, applying a coating liquid containing a silicone resin, a reaction accelerator and a solvent and drying.

The release layer may be a layer of a conventional release agent or a release layer formed on a certain sheet, for example, a release paper produced by applying a coating solution composed of a silicone resin, a reaction accelerator and a solvent to a paper and drying. For example, the surface coated with the release agent is placed on the adhesive surface of the adhesive layer.

According to the present invention, it is very important that the layers of the pressure sensitive recording sheet are combined in such a manner that the appearance of the opaque pressure clarifiable layer is different from the appearance of the portions transparentized by applying a pressure. The difference of appearance may be difference of color, shade of color and other visual properties of the surface states.

For example, in case of a pressure sensitive recording sheet composed of an adhesive layer, an opaque colored support sheet, an opaque pressure clarifiable layer and a colorless transparent release treated layer, when the color of the opaque pressure clarifiable layer and that of the opaque colored support sheet are different from each other. The color appearing at the transparentized portion is that of the opaque colored support sheet. When the transparentized portion of the opaque pressure clarifiable layer is not colorless, but colored-transparent, the color appearing at the transparentized portion is a mixed color of the opaque colored support sheet and the color of the transparentized portion of the opaque pressure clarifiable layer. Further, when there is a transparent protective layer or a release treated layer or there are both a transparent protective layer and a release treated layer and they are colored-transparent, the color appearing at the transparentized portion is a mixed color of them, and the color appearing at the untransparentized portion of the opaque pressure clarifiable layer is a mixed color of color or colors of the layers overlying the opaque pressure clarifiable layer and color of the opaque pressure clarifiable layer. Further, if adhesives used for bonding those layers are colored-transparent, the resulting appearing color is a mixed color formed by adding the color or colors of the adhesive or adhesives.

In view of the foregoing, it is very important to select the color of each layer in such a manner that the color appearing at the transparentized portion of the opaque pressure clarifiable layer is different from the color appearing at the untransparentized portion.

The above explanation is directed to the difference of color only, but it will be easily realized that any kind of visual difference is usable according to the present invention.

In general, a pressure sensitive recording sheet comprising a colorless transparent protective layer, an opaque pressure clarifiable layer and an opaque support sheet having a color different from that of the opaque pressure clarifiable layer, particularly a color well contrasting with that of the opaque pressure clarifiable layer, is preferable. The contrast of colors may be optionally selected depending upon the use of the recording sheet, for example, beautiful sense, warning, and usual indication.

Sizes and shapes of layers constituting the pressure sensitive recording sheet may be different from each

other and may be selected optionally depending upon the use of the recording sheet. Various combinations of the sizes and shapes are illustrated in the drawing though they are not limited to the illustrated ones.

In FIGS. 6, 7, 16, 17, etc., each layer has the same size and shape. In FIGS. 1A, 1B, 2, 3, etc., the transparent protective layer and the support sheet have the same size and shape. In FIG. 4 etc., the opaque pressure clarifiable layer and the support sheet have the same size and shape. In FIG. 5 etc., shape and size of the opaque pressure clarifiable layer is different from those of the support sheet. In FIGS. 1A, 1B, 2, 3, 4, 5, 10, 13, 15 and 18, the width of the opaque pressure clarifiable layer is narrower than other layers such as the support sheet. In these cases, the center portion (the opaque pressure clarifiable layer) can have a color different from that of the both edge portions. Thus it is possible to select two colors of excellent and beautiful contrast. Since it is possible to make the color appearing at the transparentized portion the same as that of the edge portion so that the color appearing at the transparentized portion can be predicted by the color of the edge portion. Furthermore, it is possible to classify the articles to which the pressure sensitive recording sheet is to be adhered, by the colors of the edge portions.

The layers and sheets constituting the pressure sensitive recording sheet may be assembled in any way depending upon its use as far as each layer or sheet is not separated away. The layers and sheets may be assembled by using adhesives or fusing or sealing. The layers and sheets can be assembled not only by assembling each layer and sheet separately prepared, but also can be produced by directly coating one or more of them.

Upon assembling it should be avoided to disturb or deteriorate the function of the recording sheet. For example, it is not allowable to use an opaque adhesive between the transparent protective layer and the opaque pressure clarifiable layer.

For the purpose of adhering the pressure sensitive recording sheet to an article though the recording sheet may be used without adhering to an article, various adhering means may be employed. For example, an adhesive layer is provided under the support sheet.

The pressure sensitive recording sheet may be commercially in a various forms such as tape type, sheet type and the like. The one of the tape type can be wound and sold in a form of a roll. When the top layer of pressure sensitive recording sheet has a release property, the roll in case of tape type having an adhesive layer can be made without using any release layer or applying a releasing treatment, and the sheet type ones can be piled without using any release sheet except a release layer for the bottom one. When the adhesive layer is made of a water soluble adhesive, in usual a release layer or sheet is not necessary.

One of preferable embodiments of the present invention is a pressure sensitive recording sheet comprising a transparent support sheet and a colored adhesive layer having a color contrasting with the color of the opaque pressure clarifiable layer.

The colored adhesive layer may be a colored pressure sensitive adhesive layer composed of a coloring agent such as benzidine yellow, phthalocyanine blue, phthalocyanine green, soluble azo dye, chromium oxide, zinc oxide and the like, and an adhesive composition, for example, rubber series adhesives such as neoprene and the like, vinyl series adhesives such as polyvinyl chloride and the like, cellulosic high polymers such as ethyl-

cellulose and the like together with an adhesiveness imparting agent such as dammar, D.O.P. and the like. The colored adhesive layer composition may be applied to the transparent support sheet by dissolving the composition in a solvent such as solvent naphtha and the like. Further, a colored water soluble adhesive layer may be employed. The colored adhesive layer may be transparent or not.

Referring to FIG. 1A, an opaque pressure clarifiable layer 2 is interposed between a transparent protective layer 1 and a support sheet 3 and transparent protective layer 1 is adhered to opaque pressure clarifiable layer 2 and support sheet 3 with an adhesive 10.

Referring to FIG. 1B, a pressure is applied to the pressure sensitive recording sheet of FIG. 1A by a stylus 20 and the pressed portion 2' of opaque pressure clarifiable layer 2 is transparentized. Thus the surface of support sheet 3 corresponding to the portion 2' can be seen through the portion 2' and this is the recording

Referring to FIG. 2, an adhesive layer 11 is applied to the support sheet 3 of the recording sheet of FIG. 1A and further a release paper 4 is applied to adhesive layer 11. Upon using, release paper 4 is removed and the recording sheet is applied to a surface of article.

Referring to FIG. 3, an opaque pressure clarifiable layer 2 is interposed between a transparent protective layer 1 and a support sheet 3 and the both edge portions of transparent protective layer 1 and support sheet 3 are adhered to each other, and an adhesive layer 11 and a release paper 4 are provided.

Referring to FIG. 4, a transparent protective layer 1, an opaque pressure clarifiable layer 2 and a support sheet 3 are closely contacted with one another and fixed to another support sheet 5 by using an adhesive agent 13 through the edge portions of transparent protective layer 1 and the bottom side of opaque pressure clarifiable layer 2, and a combination of an adhesive layer 11 and a release paper 4 is provided on the back surface of support sheet 5. According to this pressure sensitive recording sheet, the color of support sheet 5 can be different from colors of opaque pressure clarifiable layer 2 and support sheet 3 and thereby, when letters or signs are inscribed in the recording sheet, the color of the inscribed letters or signs is that of support sheet 3 and the color adjacent to the letters or signs is that of opaque pressure clarifiable layer 2 and the color at the both edge portions is that of support sheet 5 and thus, there is obtained a beautiful three color label.

Referring to FIG. 5, the pressure sensitive recording sheet is composed of an opaque clarifiable layer 2 adhered to a support sheet 3 by an adhesive agent 14. On the bottom side of support sheet 3 may be provided adhesive layer 11 and release layer 4 as shown in FIG. 2. When an opaque pressure clarifiable layer 2 is not covered with any protective layer as in this embodiment, it is preferable to use an opaque pressure clarifiable layer somewhat thicker than that covered with a protective layer, and further it is preferred that the opaque pressure clarifiable layer can sufficiently withstand external physical or chemical action.

Referring to FIG. 6, the pressure sensitive recording sheet is composed of a transparent protective layer 1, an opaque pressure clarifiable layer 2, and support sheet 3 which are assembled by using adhesive layers 15 and 16, and further an adhesive layer 11 and release layer 4. The width of each layer is the same as each other.

Referring to FIG. 7, the recording sheet is the same as that of FIG. 6 except that there are not the transpar-

ent protective layer and thereby the adhesive layer 15 and the opaque pressure clarifiable layer 2 is exposed and inscription is directly made on the opaque pressure clarifiable layer 2.

Referring to FIG. 8, the pressure sensitive layer is composed of an opaque pressure clarifiable layer 3 adhered to a transparent support sheet 4 with an adhesive layer 8, and a colored adhesive layer 5 under the transparent support sheet 4.

Referring to FIG. 9, the recording sheet is the same as that of FIG. 8 except that a release layer 6 is provided. Upon using, release layer 6 is removed and the recording sheet is adhered to an article with the adhesive layer 5.

Referring to FIG. 10, the pressure sensitive recording sheet has the same structure as that of FIG. 9 except that the width of opaque pressure clarifiable layer 3 and therefore that of adhesive layer 8 is narrower than other layers. On the upper side, there appear a color zone of the layer 3 at the center portion and color zones of the colored adhesive layer 5 at both edge portions when the transparent support sheet is colorless or color zones of a mixed color of the transparent support sheet 4 and the color adhesive layer 5 when the transparent support sheet is colored. Thus, the appearance is beautiful. Upon using, the release layer 6 is removed and the recording sheet is adhered to an article.

Referring to FIG. 11, the pressure sensitive recording sheet is composed of a transparent protective layer 1, an adhesive layer 2, an opaque pressure clarifiable layer 3, an adhesive layer 7, a transparent support sheet 4 and a colored adhesive layer 5.

Referring to FIG. 12, the recording sheet is the same as that of FIG. 11 except that a release layer 6 is provided under the colored adhesive layer 5. The release layer 6 is removed upon using and the recording sheet is adhered to an article.

Referring to FIG. 13 the layer structure of the pressure sensitive recording sheet is the same as that of FIG. 12 except that width of the opaque pressure clarifiable layer is narrower than other layers and the adhesive layer 7 is not necessary. The appearance of this recording sheet is beautiful in a similar way to that of FIG. 10.

The recording sheets as mentioned above having a release sheet are suitable for use as sheet type products.

Referring to FIG. 14, the pressure sensitive recording sheet is composed of a release treated layer 9, an opaque pressure clarifiable layer 3, an adhesive layer 8, a transparent support sheet 4 and a colored adhesive layer 5. This kind of recording sheet is suitable for use as roll type products by winding the recording sheet in a form of tape.

Referring to FIG. 15, the pressure sensitive recording sheet is composed of a release treated layer 9, a transparent protective layer 1, an adhesive layer 2, an opaque pressure clarifiable layer 3, a transparent support layer 4 and a colored adhesive layer 5 where the width of opaque pressure clarifiable layer 3 is narrower than that of other layers. The appearance of this recording sheet is similar to that of FIG. 10. This is suitable for roll type products. Naturally, the recording sheets of FIG. 14 and FIG. 15 may be piled without any additional releasing means so that they can be also used as a kind of sheet type product.

Referring to FIG. 16, the pressure sensitive recording sheet is composed of a release treated layer 5, an opaque pressure clarifiable layer 4, an adhesive layer 3, an

opaque colored support sheet 2 and an adhesive layer 1, and is suitable for roll type products:

Referring to FIG. 17, this pressure sensitive recording sheet is the same as that of FIG. 16 except that a transparent protective layer 6 and an adhesive layer 7 are additionally provided. The release treated layer 5 may be formed on the transparent protective layer 6 by treating the surface of the layer 6.

Referring to FIG. 18, the pressure sensitive recording sheet is the same as that of FIG. 17 except that the width of opaque pressure clarifiable layer 4 is narrower than other layers and thereby the adhesive layer 3 is omitted. A two-color recording sheet is obtained.

In a similar way, the recording sheet of FIG. 16 can be a two-color recording sheet by using a narrower opaque pressure clarifiable layer 3. In such case, the edge portions of the release treated layer 5 is provided directly on the opaque colored support sheet 2.

I claim:

1. A pressure sensitive recording sheet consisting of a single opaque pressure clarifiable layer composed only of non-porous unbaked polytetrafluoroethylene, a support sheet under said pressure clarifiable layer, and a layer of adhesive disposed between said pressure clarifiable layer and said support sheet, either one of said layer of adhesive or said support sheet having a color contrasting with the color of said pressure clarifiable layer.

2. A pressure sensitive recording sheet according to claim 1, wherein a transparent protective layer is provided on the opaque pressure clarifiable layer.

3. A pressure sensitive recording sheet according to claim 1 or 2, wherein a second adhesive layer is disposed under said support sheet and a release layer is disposed under said second adhesive layer.

4. A pressure sensitive recording sheet according to claim 1, wherein said layer of adhesive has a color contrasting with the color of said pressure clarifiable layer.

5. A pressure sensitive recording sheet according to claim 1, wherein said adhesive is transparent and said support sheet has a color contrasting with the color of said pressure clarifiable layer.

6. A pressure sensitive recording sheet consisting of a single opaque pressure clarifiable layer composed only of non-porous unbaked polytetrafluoroethylene, a support sheet under said pressure clarifiable layer, and a layer of adhesive disposed under said support sheet, either one of said layer of adhesive or said support sheet having a color contrasting with the color of said pressure clarifiable layer.

7. A pressure sensitive recording sheet according to claim 6 wherein said support sheet is transparent and said layer of adhesive has a color contrasting with the color of said pressure clarifiable layer.

8. A pressure sensitive recording sheet according to claim 6 wherein said support sheet has a color contrasting with that of said pressure clarifiable layer.

9. A pressure sensitive recording sheet according to claim 6, wherein a transparent protective layer is provided on the opaque pressure clarifiable layer.

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UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 4,276,334
DATED : June 30, 1981
INVENTOR(S) : TETSUO SUGIHARA

It is certified that error appears in the above—identified patent and that said Letters Patent is hereby corrected as shown below:

Column 2, line 1, after "shows", delete "that".

Column 4, line 34, "avoid" should be --avoided--;
line 51, "end" should read --and--; line 63 "phthatocyanine"
should read --phthalocyanine--.

Column 6, line 26, "lyer" should read --layer--.

Signed and Sealed this

Twenty-second Day of September 1981

[SEAL]

Attest:

GERALD J. MOSSINGHOFF

Attesting Officer

Commissioner of Patents and Trademarks



US005200268A

United States Patent [19]

Hamada

[11] **Patent Number:** **5,200,268**[45] **Date of Patent:** **Apr. 6, 1993**

[54] **STICK-ADHESIVE COMPOSITION, A STICK-ADHESIVE COMPOSITION LAYER, AND A TRANSFER SHEET WITH THE LAYER**

[76] **Inventor:** Hideo Hamada, 15-401, Kawanaka-Shinmachi, Daito-shi, Osaka 574, Japan

[21] **Appl. No.:** 678,461

[22] **Filed:** Apr. 1, 1991

[51] **Int. Cl.⁵** B32B 7/12

[52] **U.S. Cl.** 428/354; 525/123; 525/131; 428/40; 428/202; 428/343; 428/352; 428/355

[58] **Field of Search** 525/131, 123; 428/40, 428/352, 354, 355, 343, 202

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Primary Examiner—George F. Lesmes

Assistant Examiner—D. R. Zirker

Attorney, Agent, or Firm—Armstrong, Westerman, Hattori, McLeland & Naughton

[57] **ABSTRACT**

The invention relates to a transfer sheet comprising a release sheet, a stick-adhesive layer disposed on the releasable side of the release sheet and formed from the stick-adhesive composition comprising a sticking resin and a moisture-curing resin in a weight ratio of 90:10 through 20:80 based on the nonvolatile matter thereof, a main transfer layer disposed on the stick-adhesive layer, and a moisture-proofing means disposed on the main transfer layer and adapted to maintain the stick-adhesive layer in moisture-free condition.

2 Claims, 1 Drawing Sheet

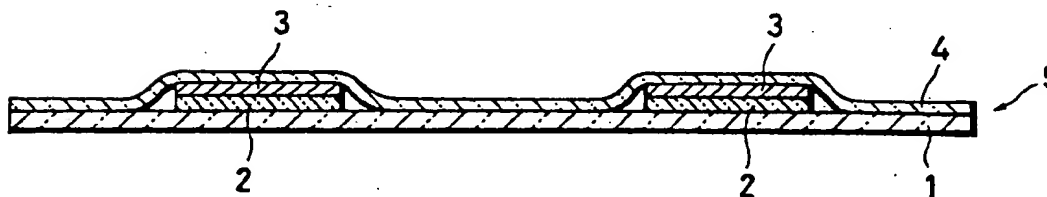
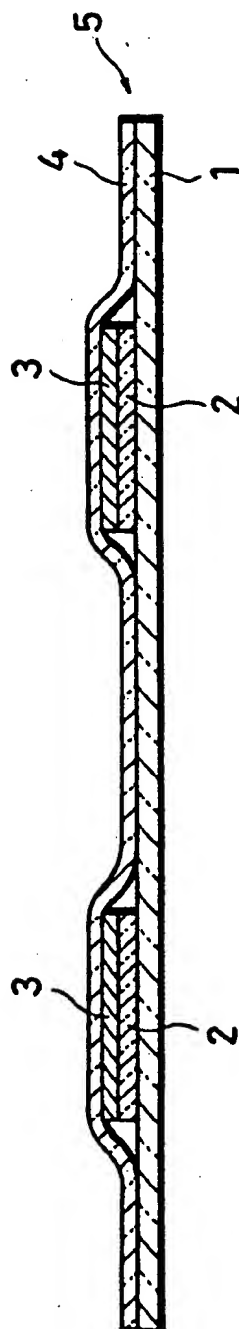


Fig. 1



STICK-ADHESIVE COMPOSITION, A STICK-ADHESIVE COMPOSITION LAYER, AND A TRANSFER SHEET WITH THE LAYER

FIELD OF THE INVENTION

The present invention relates to a stick-adhesive composition which, after application to a substrate or adherend, undergoes transition from a sticking agent to an adhesive agent with the progress of time. The invention relates, in a further aspect, to a layer formed from said stick-adhesive composition. In a still further aspect, the invention relates to a transfer sheet having the same layer.

BACKGROUND OF THE INVENTION

Sheets and the like materials each carrying a sticking layer offer the advantage that they can be stuck to substrates with so-called "fingertip ease" but are deficient in bonding strength and resistance to slip stress because of the inherent nature of the sticking agent. On the other hand, sheets and other materials each carrying an adhesive layer insure permanent bonding to adherends but require not only subsequent drying or heating but also application of a pressure over a certain time period, thus being inferior in workability.

The foregoing suggests that if a stick-adhesive composition that would undergo spontaneous transition from a sticking agent to an adhesive agent after application to any adherend should ever be developed, both the demand for ease of application and the demand for permanent adhesion could be successfully reconciled and satisfied.

Much research has been undertaken into stick-adhesive agents. By way of illustration, Japanese Kokai Tokkyo Koho No. 63-10680 discloses a stick-adhesive composition comprising a monofunctional acrylic monomer, a compound containing two or more acryloyl groups, a compound containing two or more epoxy groups and a masked epoxy-curing agent.

Japanese Kokai Tokkyo Koho No. 51-6235 discloses a stick-adhesive composition comprising a mixture of n-butyl acrylate and 2-ethylhexyl acrylate, a functional vinyl monomer and a vinyl monomer copolymerizable therewith.

Furthermore, Japanese Kokai Tokkyo Koho No. 61-66631 describes a continuous lapping method for cladding a piece of furniture or an architectural member with a decorative sheet employing a stick-adhesive composition of the urethane prepolymer type which comprises paying out an endless decorative sheet from a roll, applying said stick-adhesive composition to the reverse side of said decorative sheet in the course of its travel, drying the same and applying the sheet to the furniture or architectural member through a roll set adapted to compress the sheet against said member in registry with the surface pattern of the latter. The stick-adhesive composition employed in this method develops a pressure-sticking property within 8 hours after evaporation of the coating solvent and, thereafter, an adhesive property as the curing reaction proceeds in line with progressive absorption of moisture with or without the aid of a curing component.

However, the stick-adhesive composition proposed in Japanese Kokai Tokkyo Koho No. 63-10680 has the disadvantage that heating is required for the conversion from a sticking agent to a bonding agent. The stick-adhesive composition described in Japanese Kokai Tok-

kyo Koho No. 51-6235 is also disadvantageous in that heat curing is required after application to the substrate. This need for a post-heating procedure for said conversion from a sticking agent to an adhesive agent is not only an economic deterrent because this requires additional work after application but a serious limiting factor in the utility of the composition because it cannot be utilized in applications where heat is abhorred or in the art of transfer printing sheets which are to remain on the substrates.

The stick-adhesive composition according to Japanese Kokai Tokkyo Koho No. 61-66631 is subject to a time limit that its pressure adhesivity is available only up to 8 hours after evaporation of the coating solvent. Moreover, it can never be used for transfer sheets which are marketed in the condition preformed with a stick-adhesive layer.

It is, therefore, an object of the present invention to provide a stick-adhesive composition with which a final product can be marketed as carrying a stick adhesive layer and applied to the substrate by simple pressing as it is the case with the conventional adhesive material and which provides a permanent bond as it is simply allowed to stand and undergo spontaneous transition from a sticking agent to an adhesive agent with the progress of time after application.

It is another object of the invention to provide a stick-adhesive layer formed from such stick-adhesive composition.

It is a further object of the invention to provide a transfer sheet having said stick-adhesive layer.

SUMMARY OF THE INVENTION

The present invention provides a stick-adhesive composition which undergoes spontaneous transition from a sticking agent to an adhesive agent with the progress of time after application, which composition comprises a sticking resin and a moisture-curing resin in a weight ratio of 90:10 through 20:80 based on the nonvolatile matter thereof.

The stick-adhesive layer according to the present invention comprises a layer formed from the abovedefined stick-adhesive composition wherein said moisturecuring resin exists continually from one surface of the same layer to the other side.

The transfer sheet according to the present invention comprises a release sheet, a stick-adhesive layer disposed on the releasable side of said release sheet and formed from said stick-adhesive composition containing a sticking resin and a moisture-curing resin in a weight ratio of 90:10 through 20:80 on a nonvolatile matter basis, a main transfer layer disposed on top of said stick-adhesive layer, and a moisture-proofing means for maintaining said stick-adhesive layer in moisture-free condition.

BRIEF DESCRIPTION OF THE DRAWING

FIG. 1 is a schematic cross-section view illustrating an exemplary transfer sheet according to the invention.

DETAILED DESCRIPTION OF THE INVENTION

In accordance with the present invention, it is essential that the stick-adhesive composition contains a sticking resin and a moisture-curing resin in a weight ratio of 90:10 through 20:80 on a nonvolatile matter basis.

The sticking resin includes, inter alia, acrylic, silicone, rubber, vinyl, urethane and ultraviolet curing resins which have sticking properties particularly useful are acrylic sticking resins. Such an acrylic sticking resin can be prepared by copolymerizing a soft segment component consisting in an alkyl (about 4-12) (meth)acrylate such as butyl acrylate, 2-ethylhexyl acrylate or the like, a hard segment component consisting of a short-chain alkyl (meth)acrylate, vinyl acetate or the like, and a functional component such as an ethylenically unsaturated carboxylic acid, a hydroxy (meth)acrylate or the like in appropriate proportions.

Depending on the type of sticking resin, a tackifying agent and/or a softening agent can be used in conjunction. It is also possible to incorporate, where necessary, an aging inhibitor, filler, vulcanizing agent, crosslinking agent, colorant, antifoam, leveling agent and other additives in the aforesaid sticking resin.

The moisture-curing resin is preferably a resin for one-package NCO-terminated polyurethane adhesive compositions, for example the resins obtainable by reacting a polyisocyanate compound such as tolylene diisocyanate, diphenylmethane diisocyanate or the like with a polyol such as a polyether polyol (e.g. polyoxypropylenediol, polytetramethylene glycol ether, polyoxyethylene diol, etc.) or a polyester polyol (e.g. polyethylene adipate, polybutylene adipate, polyhexamethylene adipate, polycaprolactone, etc.). The above resin may have a urethane bond in the intermediate region of its molecule. Aside from the above-mentioned resins, it is possible to employ a moisture-curing silicone resin or an acrylic resin containing a substance which is rendered basic by exposure to moisture.

The weight ratio of said sticking resin to said moisture-curing resin is generally 90:10 through 20:80, preferably 85:15 through 40:60, and more desirably 80:20 through 50:50. If the proportion of the sticking resin is too large and that of the moisture-curing resin is too small, the desired permanent bond cannot be obtained. If the proportion of the former is too small and that of the latter is too large, the stickability of the composition is sacrificed.

It is preferable that, in the stick-adhesive layer 2 formed from said stick-adhesive composition, the moisture-curing resin is present continually between both surfaces of said layer.

If the moisture-curing resin exists in the form of an island or islands in a sea of the sticking resin, the bond strength increases as the curing reaction of the moisture-curing resin proceeds with standing time but as the time progresses further, the bond strength reaches a point of saturation and thence tends to decrease. However, since the transition from a sticking agent to an adhesive agent takes place, of course, such a layer structure can be tolerated depending on uses.

To insure that the moisture-curing resin will be distributed continually between both surfaces of the stick-adhesive layer, it is important to use a special solvent system for the stick-adhesive composition.

Thus, the solvent system to be used in the preparation of the stick-adhesive composition is preferably a system consisting of at least two solvents each dissimilar in the solubility of the sticking resin and moisture-curing resin and such that one of the solvents in which the sticking resin is more readily soluble than the moisture-curing resin is evaporated in the first place after application and the other solvent in which the moisture-curing resin

is more readily soluble than the sticking resin is evaporated in the second place.

In this arrangement, even if and when the sticking resin is precipitated during preparation of the stick-adhesive composition or with the progress of subsequent drying, the moisture-curing resin still remains in solution, with the result that the moisture-curing resin may be distributed continually between two surfaces of the stick-adhesive layer 2.

The solvents mentioned above can be selected from among, for example, hydrocarbons, esters, ethers, ketones, alcohols, polyols, nitrogen-containing solvents, sulfur-containing solvents, halogenated solvents, and solvents containing two or more functional groups (e.g. OH, ester and ether groups), and these solvents are used combinedly in suitable proportions.

Whether, in the stick-adhesive layer 2, the moisture-curing resin exists continually from one surface of the layer to the other surface or as an island or islands in the sea of sticking resin can be ascertained by a variety of techniques. For example, the internal structure of the layer can be ascertained by staining the layer with a dye which shows divergent affinities for the sticking resin and moisture-curing resin and observing the stained section of the layer under a microscope or, alternatively, by treating the stick-adhesive layer 2 after curing of its moisturecuring resin component with a solvent capable of dissolving the sticking resin and examining the residual structure of the layer.

The stick-adhesive composition of the layer 2 formed therefrom can be used in a variety of uses where it must behave as a sticking agent at application but subsequently undergo spontaneous transition to an adhesive agent. However, since one of the most pertinent uses for the composition is as a transfer sheet, the mode of application of the present invention to a transfer sheet is now described in detail.

The transfer sheet essentially comprises a release sheet 1, a stick-adhesive layer 2 disposed on the releasable surface of said release sheet, and a main transfer layer 3 disposed on said stick-adhesive layer 2. The sequence of formation of the respective layers is optional. The main transfer layer 3 may for example be a printed film, film, paper, foil, vapor-deposited layer or the like and may actually be a multi-layer structure. The transfer sheet may be previously provided with suitable cutouts.

The stick-adhesive layer 2 cures in the presence of moisture. Therefore, after fabrication of the transfer sheet, a moisture-proofing means 5 is applied so that the layer 2 may remain in moisture-free condition during storage and distribution. This moistureproofing means 5 may for example be a moisture-proof wrapping which seals the whole transfer sheet or consist in the use of a moisture-proof material for said release sheet in combination with a moisture-proof sheet releasably disposed on top of the main transfer layer 3.

Among a diversity of transfer sheets that can be provided in accordance with the present invention, the sheet is preferred in which the release sheet 1 is a moisture-proof sheet, the stick-adhesive layer 2 is a layer printed according to a pattern on the releasable surface of said release sheet 1, the main transfer layer 3 is at least one pattern-printed layer (ink layer) formed on and in substantial registry with the pattern of said stick-adhesive layer 2, and said moisture-proofing means 5 for maintaining said stick-adhesive layer in moisture-free condition is said release sheet 1 in combination with a

peelable protective sheet 4 disposed on said main transfer layer 3 in such a manner that it covers an area larger than and extending beyond the pattern of said transfer layer 3.

The release sheet 1 may be a paper or plastic sheet treated with a release agent on one side. The release agent includes, inter alia, long-chain alkyl acrylate copolymers, long-chain alkyl vinyl ester copolymers, long-chain alkyl vinyl ether copolymers, long-chain alkyl acrylamide copolymers, long-chain alkyl allyl ester copolymers, long-chain alkyl derivatives of maleic acid, long-chain alkyl ester polymers, long-chain alkyl carbamate polymers, polyethyleneimine derivatives, perfluoroalkyl compounds (inclusive of fluororesin), silicone, paraffin and so on. Particularly useful for practical purposes is silicone. Aside from these agents, the sheet as such may be made of a release material or the sheet may contain a release agent internally added in the course of manufacture of the sheet.

The pattern-printing of the releasable surface of the release sheet 1 with the stick-adhesive composition can be accomplished by a desired printing technique such as screen printing. For screen printing, the viscosity of the stick-adhesive composition is usually adjusted to 1 to 300 poises (reading of a BH rotary viscosimeter at 25° C.). The stick-adhesive layer 2 may be a single layer or a plurality of layers.

In the printing of the release sheet 1 with the stick-adhesive composition, there are cases in which the surface being printed repels the composition or an accurate pattern cannot be reproduced. In such cases, it is good practice to incorporate traces or small amounts of a surface active substance, such as silicone and other surfactants, in the stick-adhesive composition.

After formation of said stick-adhesive layer 2 on said release sheet 1, the main transfer layer (ink layer) 3 consisting of a single or plural patternprinted layers is formed in such a manner that the pattern of the layer 3 will coincide with the pattern of said stick-adhesive layer 2. The ink that can be used for this purpose is virtually unlimited and includes, inter alia, clear inks, colored inks, ultraviolet-curing ink, electron beam-curing ink and a diversity of functional inks (e.g. electrically conductive, thermo-color, fluorescent, light storage, reflective, fragrant, magnetic, OCR, desensitizing, resist, transfer printing and other inks).

Lastly, on top of the main transfer layer 3, there is provided a peelable protective sheet 4 in such a manner that it covers an area larger than and extending beyond the pattern on said main transfer layer 3.

The protective sheet 4 is advantageously a film or sheet made of a polyolefin, polyvinyl chloride, polyester, polyamide, polyurethane, polyimide, cellulose derivative, polyacetal or the like and coated either thoroughly or partially with a coating agent imparting a mild degree of sticking property depending on the intended application. It is also possible to use a plurality of protective sheets in superimposition so as to impart added body to the whole sheet. The protective sheet may also be formed by printing.

When a moisture-proof sheet is used as said release sheet 1 and, also, as said protective sheet 4, the main transfer layer 3 is sandwiched between the release sheet 1 and the protective sheet 4 so that the moisture-free condition mentioned hereinbefore can be insured. Therefore, in this instance, the release sheet 1 and the protective sheet 4 function as said moisture-proofing

means for maintaining the stick-adhesive layer 2 in moisture-free condition.

The transfer printing sheet described above has a laminar configuration of (1)/(2)/(3)/(4) and it is a usual practice to construct the layers starting with (1). However, all that is necessary is that the above configuration be finally implemented and there is no limitation on the order of formation of the layers.

To facilitate the transfer process, the bonding strength between the release sheet 1 and stick-adhesive layer 2 is set to a lowest value, that between the main transfer layer 3 and protective sheet 4 to an intermediate value and that between the stick-adhesive layer 2 and the substrate to a highest value.

Since the stick-adhesive layer 2 is sticky, the substrate may be optional in kind. Thus, the substrate includes organic and inorganic materials such as metals, plastics, coated surfaces, wood, bamboo, ceramics, paper, woven fabrics, nonwoven fabrics, leather, porcelains and so on.

The transfer procedure is as follows. First, the protective sheet 4 is released with the fingers or a jig and the laminated structure consisting of the stick-adhesive layer 2, main transfer layer 3 and protective sheet 4 is removed from the release sheet 1. Then, the stick-adhesive layer of the laminated structure is pressed against a substrate and the protective layer (4) alone is peeled off.

The present invention provides the following advantages.

Since the stick-adhesive layer 2 acts as a sticking agent, it can be stuck to any substrate, irrespective of its material, with one-touch ease.

Since the stick-adhesive layer 2 cures upon exposure to atmospheric moisture, it gradually cures as stuck to the substrate after it has contacted the air in application.

Where, in the stick-adhesive layer formed from the stick-adhesive composition of the invention, said moisture-curing resin exists continually from one surface of the layer 2 to the other surface, progression of the curing reaction results in a firm permanent bond between the substrate and the main transfer layer.

Thus, in the present invention, the sticking resin and moisture-curing resin constituting the stick-adhesive layer 2 can fully display their respective characteristic properties. In other words, the sticking resin plays its role completely at application to the substrate and, after application, the moisture-curing resin discharges its duty completely. Furthermore, while the moisture-curing resin is playing its role, the sticking resin assures a sustained contact between the composition and the substrate, with the result that the desired adhesion can be materialized with intimate contact maintained therebetween. In addition, whereas the moisture-curing resin by itself is inherently deficient in low-temperature impact resistance, the presence of the sticking resin in the composition of the invention obviates the problem of this deficiency in low-temperature impact resistance.

The transfer sheet having the stick 2 according to the present invention can be launched into market distribution as such, namely carrying the stick-adhesive layer 2, provided that the aforesaid moisture-proofing means 5 is provided for maintaining the stick-adhesive layer 2 in moisture-free condition (for example, the interposition of the stick-adhesive layer 2 between the release sheet 1 and the protective sheet 4), for such moisture-proofing means 5 prevents curing of the moisture-curing resin for a period of several months to more than a year. The user

may purchase this transfer sheet and apply it to the substrate he desires with one-touch ease.

When the stick-adhesive layer 2 of the present invention is applied to a transfer sheet, the curing reaction of the moisture-curing resin in the stick-adhesive layer 2 proceeds from the edge of the transferred layer, thus insuring a high anchoring effect with respect to the substrate at the edges where the highest adhesive strength is required.

The following examples are further illustrative of the invention and should by no means be construed as defining the metes and bounds of the invention.

EXAMPLE 1

A sticking resin solution prepared by dissolving n-butyl acrylate-vinyl acetate (7:3 by weight) copolymer in ethyl acetate-toluene (resin content 40%, viscosity 5000 cps/25° C.) was mixed with a moisture-curing resin solution prepared by dissolving a urethane resin in ethyl acetate-toluene (resin content 30%, viscosity 1000 cps/25° C.) in a predetermined ratio to make a total of 100 parts. The mixture was then diluted with 50 parts of ethylene glycol monoethyl ether acetate which is a good solvent for both the sticking resin and moisture-curing resin to provide Composition B.

On the other hand, the above-mentioned sticking resin solution and moisture-curing resin solution were mixed in a predetermined ratio to make a total of 100 parts. This mixture was diluted with 50 parts of a solvent mixture consisting of 70% of diethylene glycol ethyl ether acetate, which is a good solvent for both the sticking resin and moisture-curing resin, and 30% of dimethyl sulfoxide, which is a good solvent for said moisture-curing resin but a poor solvent for said sticking resin to provide Composition A.

Using a bar coater with a clearance of 0.1 mm, each of Composition A and Composition B was coated on releasable side of a polyester release sheet 1. After 10 minutes of drying at 80° C., a protective sheet 4 comprising a polyvinyl chloride film carrying a weakly sticking layer was superimposed and pressed on the coated side. This laminate was cut to a width of 5 mm and, after the protective sheet 4 was peeled off, applied to the untreated side of a 125 μ m thick polyester substrate film. The assembly was allowed to stand and the peeling strength was sequentially measured. The results are set forth below. The mixing ratios shown are by weight of the sticking resin solution/moisture-curing resin solution.

(a) When Composition A was used

Time in days	Mixing ratio				
	100/0	90/10	80/20	70/30	60/40
Immediately after application	330	280	300	330	350
4	300	300	360	360	380
11	330	340	390	400	430
18	330	350	420	430	490
25	330	380	470	490	480
32	310	400	465	490	480
39	310	430	470	480	480
46	310	430	470	480	485
53	320	430	460	480	470

*Each figure represents peeling strength (g/5 mm).

(b) When Composition B was used

Time in days	Mixing ratio				
	100/0	90/10	80/20	70/30	60/40
Immediately after application	330	280	330	370	400
4	300	320	340	420	380
11	330	320	360	410	360
18	330	350	420	400	320
25	330	370	360	360	340
32	310	390	360	340	320
39	310	410	340	350	320
46	310	340	340	360	310
53	320	320	300	320	310

*Each figure represents peeling strength (g/5 mm).

In both cases (a) and (b), the mode of attachment at the mixing ratio of 100/0 was by sticking irrespective of standing time and with regard to the mixing ratios of 90/10, 80/20, 70/30 and 60/40, the mode of attachment was still by sticking up to around day 20 but was substantially by adhesion after day 30.

Microscopic observation of the stained section of the cured stick-adhesive layer 2 revealed that whereas the moisture-curing resin was found to be dispersed in the insular fashion in the case of (b) where Composition B was used, it existed continually from one surface of the stick-adhesive layer 2 to the other surface in the case of (a) where Composition A was used.

EXAMPLE 2

Among a series of Composition A prepared in the same manner as Example 1, the composition corresponding to the mixing ratio of 75:25 (sticking resin/moisturecuring resin, by weight) was coated on the releasable side of a polyester release sheet 1 by means of a bar coater with a clearance of 0.1 mm, followed by drying at 80° C. for 10 minutes. A protective sheet 4 consisting in a polyvinyl chloride film carrying a weakly sticking layer was then laminated.

The whole laminate was cut to a width of 5 mm and stuck to various substrates, viz. stainless steel sheet, rigid polyvinyl chloride plate, polymethyl methacrylate plate, polystyrene plate and polyurethane-coated plate (which are referred to as Substrates 1, 2, 3, 4 and 5 respectively) Each assembly was then allowed to stand and the peeling strength was sequentially determined.

The results are set forth below.

Time in days	Substrate				
	1	2	3	4	5
1	260	360	370	380	220
7	310	390	415	405	315
14	330	420	440	430	380
25	380	450	460	450	420

*Each figure represents peeling strength (g/5 mm).

In contrast, in cases where the sticking resin solution alone was used, the peeling strength was about 120 g/5 mm for Substrate 1, about 220 g/5 mm for Substrate 2, about 245 g/5 mm for Substrate 3, about 220 g/5 mm for Substrate 4 and about 210 g/5 mm for Substrate 5, irrespective of the passage of time.

EXAMPLE 3

FIG. 1 is a schematic cross-section view showing an exemplary transfer sheet according to the present invention.

On the releasable side of a 125 μ m thick polyester release sheet 1, a pattern was screen-printed with Composition A wherein the weight ratio of sticking resin to moisture-curing resin was 70:30, followed by drying to provide a stick-adhesive layer 2.

This stick-adhesive layer 2 was screen-printed with a colored ink in exact registry with the pattern of the layer 2 followed by drying to provide a main transfer layer (ink layer) 3. (Incidentally, this main transfer layer 3 is formed as multiple layers in many cases).

The above procedure gave a transfer layer consisting of the stick-adhesive layer 2 and main transfer layer 3 on the release sheet 1. Then, a protective sheet 4 comprising a polypropylene film carrying a weakly sticking layer was superimposed on the above transfer layer and the assembly was passed with a pair of rubber rollers to provide an integral sheet.

The transfer sheet (transfer printing sheet) thus obtained has a laminar configuration of (1)/(2)/(3)/(4). Then, a half-cut is made between patterns on the protective sheet 4 side to provide individual transfer printing sheets.

From this transfer printing sheet, the laminate of (2)/(3)/(4) was removed from the release sheet (1) by holding one end of the protective sheet (4) with fingers and the stick-adhesive layer 2 is applied against an ABS resin plate and pressed down with fingers a few times. Then, the protective sheet 4 was peeled off, whereupon only the pattern of the transfer layer consisting of 2 and 3 remained on the ABS plate. Thus, transfer printing could be consummated with fingertip ease. The change in peeling strength with time was investigated using a large number of individual transfer printing sheets.

The transfer-printed ABS resin plates were allowed to stand and serially subjected to the peeling test described above, 5 sheets at each time. The results were as follows.

Time in days	Peeling strength (g/5 mm)
Immediately after printing	360
Day 1	380
Day 4	420
Day 11	430
Day 18	440
Day 25	435
Day 39	435
Day 46	430
Day 90	435

Observation of the peeling state revealed that whereas the mode of attachment was by sticking up to day 3, it was substantially by adhesion after day 5.

EXAMPLE 4

The transfer printing sheets of Example 3 were allowed to stand indoors, without packaging, for 1, 2, 3 and 6 months.

Using the transfer printing sheets left standing for the above-mentioned time periods, transfer to the untreated side of a polyester film was carried out in the same manner as Example 3. The peeling test was performed after a further standing time of 15 days.

The results were as follows.

Time before transfer	Time after transfer	Peeling strength (g/5 mm)
1 Months	15 Days	430
2 Months	15 Days	430
3 Months	15 Days	420
6 Months	15 Days	420

What is claimed is:

1. A transfer sheet comprising a release sheet, a stick-adhesive layer disposed on the releasable side of said release sheet and formed from a stick-adhesive composition comprising a sticking resin capable of functioning as a sticking agent for sustained contact to a substrate and a moisture-curing resin curable upon exposure to atmospheric moisture to provide an adhesive to a substrate, the resins of the stick-adhesive being in a weight ratio of sticking resin to moisture-curing resin in the range of 90:10 through 20:80 based on nonvolatile matter thereof, a main transfer layer disposed on said stick-adhesive layer opposed from said release sheet, and a moisture-proofing means disposed on said main transfer layer and adapted to maintain said stick-adhesive layer in moisture-free condition, said release sheet is a moisture-proof sheet, said stick-adhesive layer is a pattern-printed layer formed on the releasable side of said release sheet, said main transfer layer is at least one pattern-printed layer having a pattern in registry with that of said stick-adhesive layer, and said moisture-proofing means for maintaining said stick adhesive layer in moisture-free condition comprises said release sheet in combination with a peelable protective sheet disposed on said main transfer layer in such a manner that it covers an area larger than and extending the pattern of said main transfer layer.

2. The transfer sheet of claim 1 wherein the moisture-curing resin extends continually from one surface of the stick-adhesive layer to the other surface.